

Spins in the Vortices of a High Temperature Superconductor

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Neutron scattering is used to characterise the magnetism of the vortices for the optimally doped high-temperature superconductor $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x = 0.163$) in an applied magnetic field. As temperature is reduced, low frequency spin fluctuations first disappear with the loss of vortex mobility, but then reappear. We find that the vortex state can be regarded as an inhomogeneous mixture of a superconducting spin fluid and a material containing a nearly ordered antiferromagnet. These experiments show that as for many other properties of cuprate superconductors, the important underlying microscopic forces are magnetic.

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Many of the practical applications of type-II superconductors, rely on their ability to carry electrical currents without dissipation even in magnetic fields greater than the Meissner field, below which the superconductor excludes magnetic flux entirely. At such high magnetic fields, superconductors are in a mixed state or 'vortex lattice', comprising an array of cylindrical inclusions (vortices) of normal material in a superconducting matrix. Vortex lattices have two magnetic aspects. The first is that there are magnetic field gradients due to the inhomogeneous flux penetration - each vortex allows a magnetic flux quantum to penetrate and the magnetic field decays from the vortex center into the superconductor over a distance of order the London length λ (1). λ is the depth beyond which the superconductor excludes small fields, and is typically between 100 and 1000 nm. The second is that the electron spins in the non-superconducting cores should no longer be paired coherently (as they are in the superconducting state). The length scale for this microscopic magnetic effect is the radius ξ of the Cooper pairs which underlie the phenomenon of superconductivity. ξ generally ranges from 100 nm - common for conventional, low transition temperature (T_c) superconductors - down to nearly a nm, which is found for the high- T_c copper oxides. Most magnetic measurements of the flux lattice state, including images from neutron diffraction and microscopy of magnetic nanoparticles deposited on samples threaded by vortices, are sensitive primarily to the mesoscopic field gradients characterized by λ (2-5). Much less is known about the microscopic magnetism of the vortices. The associated spin correlations and dynamics are important because they mirror the internal structure of the vortices and in superconductors with strong magnetic interactions, are likely to dominate vortex state energetics and thermodynamics. Thus motivated, we have performed an experiment which images the spin correlations in the vortex state of the simplest high-temperature superconductor, $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. The key finding is that the vortex state for our optimally doped sample ($x = 0.163$, superconducting transition temperature $T_c = 38.5$ K) has much stronger tendencies towards magnetic order than either the normal or superconducting states.

We used inelastic neutron scattering to measure χ'' (the Fourier transform of the two-spin correlations divided by the Bose factor) as a function of momentum and energy. The superconducting CuO_2 planes of our sample were placed in the horizontal scattering plane of a neutron scattering spectrometer, and the magnetic field H was applied perpendicular to these planes in the vertical direction¹. A sliver of one crystal was used for magnetotransport measurements, and these were combined with earlier data for $x = 0.17$ (9) to establish the $H - T$ phase diagram (Fig. 1A). The electrical resistance vanishes below an irreversibility line (10) which is a very rapid function of applied field, so that even for fields well below the upper critical field H_{c2} (defined here as the field at which non-zero resistivity is first detected), the vortex lattice required for macroscopic superconducting phase coherence and perfect conductivity does not occur until T is well below $T_c(H = 0) = 38.5$ K, the zero field transition temperature.

$\text{La}_{1.837}\text{Sr}_{0.163}\text{CuO}_4$ is characterised by spin fluctuations occurring at a quartet of x -dependent characteristic wavevectors given by $\mathbf{Q}_\delta = (\frac{1}{2}(1 \pm \delta), \frac{1}{2})$ and $(\frac{1}{2}, \frac{1}{2}(1 \pm \delta))$ with $\delta = 0.254$ (11,12) (Fig. 1B). Superconductivity has several effects on the magnetic fluctuations, the most pronounced of which is that an energy gap, Δ , appears in the spectrum (6,13) (Fig. 2A). On cooling from $T_c=38.5$ K to 5.5 K in zero field, the normal state continuum is eliminated below $\Delta=6.7$ meV. Application of a 7.5 T field fills the gap at base temperature with a spectrum whose amplitude is little different from that seen in the same energy range in zero field at T_c . This result means that the vortex state for a field far below the upper critical field $H_{c2} \approx 62$ T (9), where the vortex cores presumably occupy a small volume fraction of the material ($H/H_{c2}=12\%$), displays low-frequency magnetic fluctuations of roughly the same strength as the ungapped normal state. The difference plot (Fig. 2B) shows that the field-induced signal peaks at 4.3 ± 0.5 meV, and dwindles to zero as E approaches either 0 or Δ . This characteristic energy is approximately half that of the normal state indicating that the fluctuations in the field are two times slower and have a greater tendency towards antiferromagnetic order.

¹The single crystal samples were described previously (6), and 11 of them (with a total weight of 25 grams) were mutually aligned to within ± 0.6 and ± 3.9 degrees in directions parallel and perpendicular to the CuO_2 basal planes respectively. The resulting sample was placed on the cold finger of a variable temperature insert in a split coil superconducting magnet, which in turn was installed on the sample table of the RITA (TAS6) cold neutron spectrometer at DR3, Risø National Laboratory (7,8).

To understand the spatial nature of the field-induced sub-gap fluctuations, we have performed scans for fixed energy transfer as a function of wavevector along the solid black trajectory shown Fig. 1B. At low temperatures and energies below the gap ($T = 6.6$ K, $E=2.5$ meV) sharp peaks are observed in a field of $H=7.5$ T (Fig. 3A) at the same wavevectors where the normal state response is maximal (Fig. 3B). We also investigated the magnetic correlations just above the spin-gap at 7.5 meV and find that the field has no discernable effect on the magnetic correlations at this energy (Fig. 3C and D).

Closer examination of Figs 3A-D yields a wealth of quantitative information about the microscopic magnetism of the vortex state. First, the preferred periodicity, derived from the peak positions Q_δ of the magnetization density, is $1/\delta = (3.93 \pm 0.09)a_0$, where $a_0 = 3.777$ Å is the Cu^{2+} - Cu^{2+} separation. This value is indistinguishable from the periodicity found for the normal state. Second, the scattering profile is slightly different from that measured at 38.5 K for $H = 0$, in that the peaks seem sharper in the vortex state even though the scattering between the peaks has the same amplitude when scaled to the peaks. Indeed the peaks are as sharp as the instrumental resolution permits, implying that the principal (period $1/\delta$) magnetization oscillations in the vortex state are coherent over distances $l_v > 20a_0$, to be compared to distances $l_n = (6.32 \pm 0.22)a_0$ for the normal state (6). For comparison, the lattice constant for a well-formed (Abrikosov) vortex state is $a_V = (2\Phi_0/\sqrt{3}H)^{1/2}$ where the magnetic flux quantum $\Phi_0 = 2067$ T(nm)². At 7.5 T, $a_V = 47.2a_0$, a number much larger than $1/\delta$, but potentially similar to l_v .

We have also measured the temperature dependence of the field-induced response. Fig. 4 shows electrical resistivity as well as neutron data, collected with wavevector and energy fixed at Q_δ and 2.5 meV respectively. At $H = 0$, the neutron signal undergoes a sharp drop starting at T_c (Fig. 4C), (which is where the transition to zero resistance also occurs (Fig. 4A)) and it dwindles into the background below 15 K. A field of $H = 7.5$ T has a large effect on the temperature evolutions of both the resistivity and neutron intensity. The resistivity descends steadily as temperature decreases between T_c and 30 K (Fig. 4A), and does not have its final inflection point, as measured by $d\rho_{ab}/dT$ (Fig. 4B) until 25K, which is also where irreversibility sets in. This inflection point has been found (14) to coincide with the drop in the magnetization associated with the freezing transition of vortices - above the freezing point, the imposed current loses energy via vortex motion, while below, the vortices are pinned and the current is dissipationless. The corresponding magnetic neutron scattering signal (Fig 4C), which is slightly suppressed at $T > T_c$, remains close to its normal state value for $T_c > T > 25$ K, and undergoes a sharp decline below 25 K. Thus, our spin signal, a microscopic probe of vortices, tracks a macroscopic measure - namely the electrical resistance - of vortex freezing, which in layered materials such as the cuprates can occur well below T_c even for $H \ll H_{c2}$, the upper critical field (15).

How can our microscopic results be connected to the bulk data? In the vortex fluid state for $T > 25$ K, all Cu^{2+} sites are visited occasionally by vortices and then depart. While at the sites, they establish a decaying (in time) magnetization density wave, the quantity which our experiment is sensitive to. Thus all Cu^{2+} sites would have some memory of visits by vortices. When the inverse residence time τ^{-1} of a vortex at any site approaches the frequency of the measured spin fluctuations the fraction of sites with such memory will begin to significantly exceed the fraction H/H_{c2} of sites covered by vortices at a given instant. The resistivity data in Fig. 4A yield the crude estimate 2.5 meV for $\hbar\tau^{-1}$ at 30 K (16), which happens to coincide with $\hbar\omega$ in Fig. 4C. As T is cooled below 25 K, the vortices become pinned via a combination of their mutual interactions and intrinsic disorder, so that they are always present at certain sites and never present at others. The outcome is then that subgap magnetization fluctuations occur only near the relatively small fraction of sites where vortices are pinned, with the result that the magnetic response is correspondingly reduced.

Although observing vortex freezing via the electron spin correlations is unprecedented, an even more fascinating phenomenon occurs below 10 K. Here, the decline of the signal below the freezing transition is reversed, resulting in a susceptibility approximately equal to the normal state χ'' . Macroscopic measurements (9) do not indicate any changes in the vortex order or dynamics. Therefore, while they can plausibly account for the abruptly falling signal near 25 K, such changes cannot be responsible for the rising signal below 10 K. We conclude that the low- T increase can only follow from changes in the magnetism of the frozen vortex matter, and speculate that its most likely cause cannot be the relatively large magnetic interactions we suspect exist within individual vortices, but rather the weaker interactions between spins in different vortices that become relevant only at low- T .

Our data show that a modest field induces extraordinary subgap excitations in the optimally doped

high- T_c superconductor $\text{La}_{1.837}\text{Sr}_{0.163}\text{CuO}_4$. There are several possible origins for such excitations. The first are the quasiparticles inhabiting the vortex cores, which in conventional superconductors are simply metallic tubes with finite-size quantization of electron orbits perpendicular to the tube axes (17,18). The second, felt to be responsible for the \sqrt{H} low- T specific heat in d -wave superconductors, is due to the nodal quasiparticles whose energies are Doppler-shifted by the supercurrents around the vortices (19). The third is that the cores are small antiferromagnets, but that because of finite size quantization, and the weak magnetic interactions between planes as well as between vortices within the same planes, the antiferromagnetic correlations are dynamic and so are characterized by finite oscillation frequencies and relaxation rates. The first are excluded because the signal which we measure is comparable to that found in the normal state, giving a superconducting to normal state signal ratio which is much larger than the volume fraction H/H_{c2} occupied by vortices in such models. The second has an analogous difficulty with the low- T specific heat C , which does appear to follow the d -wave prescription for our samples (20), in that the ratio of the low temperature (for 7.5 T) and paramagnetic phase Sommerfeld constants C/T is 15 %, and is therefore much less than the 100 % ratio of field-induced low- T to zero-field paramagnetic signals measured with the neutrons. This leaves us with the third option, where we imagine the vortex state as an inhomogeneous mixture not only of paramagnetic and superconducting regions, but as a magnetically inhomogeneous mixture as well. The superconducting regions have a well-defined spin gap, while the paramagnetic regions contain fluctuations towards long period magnetic order. This picture accounts better for the observed subgap spectral weight than the other two scenarios. Specifically, we estimate that the net subgap weight, (placed in absolute units using normalization to phonon scattering (21)) integrated over energy and reciprocal space, corresponds to $0.05\mu_B^2/\text{Cu}^{2+}$, which is remarkably close to $H/H_{c2}\mu_{2D,S=\frac{1}{2}}^2 = 0.044\mu_B^2$, the product of the volume fraction occupied by the vortices and the square of the ordered moment $\mu_{2D,S=\frac{1}{2}} = 0.6\mu_B$ found in insulating two-dimensional $S=1/2$ Heisenberg antiferromagnets (22). In other words, the ordered moment which for the model insulator appears as an elastic Bragg peak, becomes a fluctuating moment manifested in the inelastic subgap peak for the vortex state of the superconductor.

While the simple picture of inclusions of finite-size vortices with large spin density wave susceptibilities accounts for many of our observations, the material in a field cannot be simply visualized as a superconductor perforated by an array of independent, nearly antiferromagnetic cylinders with diameter given by the pair coherence length. First, the spin density period is of order the pair coherence length ξ , and the magnetic correlation length is substantially longer than ξ . Second, the magnetic field also induces broad scattering between the incommensurate peaks, with a characteristic length scale of order $a_0 \ll \xi$. Third, as described above, the low- T rise in χ'' is difficult to explain without invoking weak interactions between the spins in 'separate' vortices. The observations together show that the spins in the vortices are correlated over a variety of length scales from the atomic to the mesoscopic. The most natural explanation is that the vortices themselves are highly anisotropic objects, or at the very least, have a highly anisotropic effect on the spin correlations in the intervening superconducting region. Such anisotropy is quite consistent with a d -wave pairing state, although it has not been found in scanning probe images of vortices of high- T_c superconductors (23,24).

We have measured the microscopic spin correlations associated with the vortex state in the optimally doped single-layer high- T_c cuprate $\text{La}_{1.84}\text{Sr}_{0.16}\text{CuO}_4$. We discover that on cooling in a modest field, low energy spin fluctuations are suppressed not near the zero-field transition, but at the irreversibility line below which the superconductor is in a true zero resistance state. This links the development of the spin gap more to superconducting phase coherence - required for zero electrical resistance - throughout the sample than to local pairing. A second discovery is that at low temperatures, the vortex matter exhibits a rising tendency towards the magnetic order found for the 'striped' state (25-27) with $x=1/8$. This notion is in broad agreement with theoretical ideas (28-30) and implies that in the H - T plane, the critical line separating frozen from fluid-like 'vortex' states may actually mark mesoscopic phase separation - or 'gellation' - into a nearly magnetic insulating vortex network bathed in a superconducting quantum spin fluid.

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FIG. 1. Phase diagram and wavevector map of $\text{La}_{1.837}\text{Sr}_{0.163}\text{CuO}_4$. (A) Shows the red irreversibility line in the $H - T$ plane which separates the resistive normal/vortex fluid state from the superconducting state. Red circles come from the magneto-transport measurements (shown in Fig. 4A) and mark the temperatures where non-zero resistivity is first detected for a given field. We also show the data (red squares) of Ando and coworkers (9) for an $x=0.17$ sample. The blue arrow represents the trajectory of the $H = 7.5$ T temperature scan shown in Fig. 4C. (B) shows reciprocal space for the superconducting CuO_2 planes of $\text{La}_{1.837}\text{Sr}_{0.163}\text{CuO}_4$, as probed by our neutron scattering measurements. Spin fluctuations are observed at a quartet of incommensurate wavevectors indicated by the red dots. The solid black arc shows the wavevectors measured in a typical constant-energy scan (see Fig. 3) and the green ellipse represents the instrumental resolution. The magnetic field was applied perpendicular to the CuO_2 planes, and the blue dots indicate the reciprocal lattice associated with the $H = 7.5$ T vortex state.

FIG. 2. Constant-wavevector scans plotted as functions of energy. The energy resolution is 0.41 meV full-width-at-half-maximum. (A) shows the magnetic susceptibility χ'' , measured at the incommensurate peak \mathbf{Q}_δ , in zero applied field for both the superconducting state (red circles) and the normal state (red triangles). χ'' was also measured in a magnetic field of $H = 7.5$ T at $T = 7.7$ K (blue circles). The blue line through the data in (A) corresponds to the damped harmonic oscillator $E_o^2 E \gamma / ((E^2 - E_o^2)^2 + E^2 \gamma^2)$ (with $E_o = 4.3 \pm 0.5$ meV and $\gamma = 4.3 \pm 0.2$ meV), which models the magnetism of the vortices plus the gapped form described in Ref. (6), to account for the remaining superconducting signal. The dashed red line is the form from Ref. (6) alone and describes the gapped spin-fluid-like response of the superconductor at $H = 0$ T. The dotted red line is the quasi-elastic response $E\Gamma/(E^2 + \Gamma^2)$ (with $\Gamma = 9$ meV), that was used previously to account for the normal state signal (6). (B) shows the difference between the $H = 7.5$ T and $H = 0$ results at low T . The solid blue line is simply the difference between the blue and dashed red lines in (A).

FIG. 3. Constant-energy scans plotted as a function of wavevector along the black trajectory shown in Fig. 1B. (A) shows the susceptibility measured for $T = 6.6$ K, below the energy gap. The data are sums of scans for $E = 1.5$, 2.5 and 3.5 meV. In zero field (red circles) the susceptibility is completely suppressed by superconductivity and application of a 7.5 T field (blue circles) induces a subgap signal. For comparison the normal state susceptibility is shown in (B). (C) and (D) give the susceptibility above the energy gap at $E = 7.5$ meV, in both the normal and superconducting states respectively. The lines in all frames except (A) are the resolution-corrected Sato-Maki lineshape (31); for $H = 0$, the width parameters were fixed at the values established from the higher resolution data of Ref. (6). The solid blue line in (A) is the fit to the normal state data from (B), scaled to match the peak amplitudes, while the dashed blue line consists of two peaks representing the resolution of the instrument.

FIG. 4. Temperature-dependent electrical transport and neutron data. (A) Shows the in-plane resistivity data collected at a variety of fields from $H = 0$ to 9 T. (B) gives the derivative of the in-plane resistivity with respect to temperature at $H = 7.5$ T, which is the field employed in the neutron scattering experiment. (C) shows the magnetic susceptibilities χ'' at $\mathbf{Q} = \mathbf{Q}_\delta$ and below the energy gap at $E = 2.5$ meV, for $H = 0$ (red circles) and 7.5 T (blue circles).







